

Interplay of ferromagnetism and static band Jahn-Teller distortion in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$

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Abstract : The manganite system $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ is a ferromagnetic metal in presence of static Jahn-Teller distortion. The system is described by an electronic Hamiltonian containing the hopping terms as well as the ferromagnetic interaction term arising out of the same itinerant e_g electrons of Mn ion in presence of lattice distortion. The one particle Green's functions of the electrons are calculated by Zubarev technique using equations of motion method. The magnetization and lattice strain are calculated and solved self-consistently taking into account the impurity and chemical potential. The interplay between ferromagnetism and lattice distortions is discussed in this communication.

Keywords : Electron-electron interaction, magnetically ordered materials, exchange and super exchange.

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1. Introduction

The essential physics of the “colossal magnetoresistance” materials $\text{Re}_{1-x}\text{A}_x\text{MnO}_3$ (here Re is a rare earth such as La and A is a divalent element such as Sr or Ca) is the interplay between a strong electron-phonon coupling and the “double exchange” effect of spin alignment on electron kinetic energy [1]. In the interesting doping range $0.2 \leq x \leq 0.5$, $\text{Re}_{1-x}\text{A}_x\text{MnO}_3$ is a ferromagnetic metal at low temperature and a poorly conducting paramagnet at high temperature; the paramagnetic-ferromagnetic transition occurs at an x dependent transition temperature $T_c(x) \sim 300$ K and is accompanied by a large drop in resistivity [2,3]. The colossal magnetoresistance which has stimulated the recent activities in these materials is observed for temperature near $T_c(x)$ [4]. The electronically active orbitals are the Mn $3d$ orbitals and the mean number of d electrons per Mn is $4 - x$. The cubic anisotropy and Hund's rule coupling are so strong that three of the electrons go into the crystal field split t_{2g} core states and make up an electrically inert core spin S_c of magnitude $3/2$ and the remaining $(1 - x)$ electrons

go into conduction band of width ~ 2.5 eV made mostly of outer e_g orbitals. The e_g electrons are aligned to the core states by a Hund's rule coupling J_H which is believed to be large [2,3].

The large J_H modulates the hopping of an outer-shell electron between two Mn sites by the relative alignment of the core spins, being maximal when the core spins are parallel and minimal when they are antiparallel. Also, electron hopping promotes ferromagnetic order. This phenomenon, called double exchange [5–7], has been widely regarded [8–11] as the only significant physics in the regime $0.2 \leq x \leq 0.5$. However, it was previously shown [12] that double exchange alone cannot account for the very large resistivity of the $T > T_c$ phase or for the sharp drop in resistivity just below T_c , and was suggested that the necessary extra physics is a strong electron-phonon coupling due in part to a Jahn-Teller splitting of the Mn e_g states. The cubic-tetragonal phase transition observed for $0 \leq x \leq 0.2$ is known to be arising due to a frozen-in Jahn-Teller distortion with long-range order at the wave vector (π, π, π) .

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2. Formalism

The present model considers the itinerant e_g electrons of Mn atoms disregarding the anti-ferromagnetically ordered localized t_{2g} electrons. The strong Hund's coupling aligns the localized spins in the t_{2g} state. Because of the ionic character of the material, there exists strong electron correlation among the itinerant electrons. The ferromagnetism is induced in the conduction e_g electrons because of their interaction with the localized spins. In the simplified model under consideration, we assume the existence of a direct Heisenberg type exchange interaction between the itinerant electrons to be responsible for their ferromagnetic state. Furthermore, calculations are performed in the mean field approximation accounting for the ferromagnetic exchange interaction in the Ising limit. Since the e_g band is doubly degenerate a band Jahn-Teller distortion is invoked to remove the degeneracy. The aim of the calculation is to study the interplay between the transitions from the paramagnetic to the ferromagnetic state and the structural transition induced by the band Jahn-Teller distortion. The different terms in the model Hamiltonian are detailed below. The band Hamiltonian

$$H_i = \sum_{k,\sigma} (\epsilon_0(k) - \mu) (c_{1k,\sigma}^\dagger c_{1k,\sigma} + c_{2k,\sigma}^\dagger c_{2k,\sigma}) \quad (1)$$

represents the non-interacting electronic energy in a two-fold degenerate (e_g) band with single particle energy $\hat{I}_0(k)$ and chemical potential μ . The operators $c_{ak\sigma}^\dagger$ ($c_{ak\sigma}$) being the creation (annihilation) operator of the electron in the band a of spin s and energy $\hat{I}_0(k)$ ($a = 1, 2$; is the orbital index).

The Heisenberg exchange interaction between the spins of the e_g electrons at the i -th and j -th atomic sites is

$$H_M = -\sum J_{ij} S_i \cdot S_j. \quad (2)$$

The Ising limit was obtained by taking the z -component of the interaction $S_i \cdot S_j$ and then using the mean field approximation like $S_i^z \langle S_j^z \rangle + S_j^z \langle S_i^z \rangle - \langle S_i^z \rangle \langle S_j^z \rangle$, the Hamiltonian H_M can be written as

$$H_M = \frac{JM}{2} \sum_{\alpha,k} [c_{\alpha k \uparrow}^\dagger c_{\alpha k \uparrow} - c_{\alpha k \downarrow}^\dagger c_{\alpha k \downarrow}], \quad (3)$$

where the ferromagnetic magnetization $M = -J \langle S^z \rangle$ and J is the effective interaction energy. The electron hops in the ferromagnetic metallic state. The exchange energy per atom is comparable to the kinetic energy and is taken to ~ 1 eV.

The coupling between the electron density in a degenerate electron band with static elastic strain mode is in general, of the form

$$H_{e-L} = \sum_{i,j,l,m} G_{ij,lm} \hat{e}_{ij} \hat{\rho}_{lm}, \quad (4)$$

where $\hat{\rho}_{lm} = \sum_{k,\sigma} c_{lk\sigma}^\dagger c_{lk\sigma}$, the electron density of the unperturbed degenerate band, \hat{e}_{ij} the static strain tensor (corresponding to the zone centre $q = 0$ phonons) and G is the strength of the electron lattice interaction. In presence of tetragonal distortion, \hat{e}_{ij} is a constant and the eq. (4) simplifies to

$$H_{e-L} = \sum_{k,\sigma} G e (c_{1k,\sigma}^\dagger c_{1k,\sigma} - c_{2k,\sigma}^\dagger c_{2k,\sigma}). \quad (5)$$

Therefore, the interaction of the electrons in the degenerate conduction band with the lattice acts as external perturbation to the free electrons in the band which tries to create a population difference between the two bands. As the population difference increases, the strain builds up in the system resulting in a splitting of the single degenerate band into two with band energies $\epsilon_{\pm 2}(k) = \epsilon_0(k) \pm Ge$, provided there is a net gain in the electronic energy due to the redistribution of electrons between the split subbands in comparison to the cost in the elastic energy. Under this situation there are two non-degenerate bands separated by a gap of magnitude $2Ge$.

Hence the manganite system exhibiting Jahn-Teller static lattice distortion and ferromagnetism can be described by the total Hamiltonian

$$H = H_i + H_M + H_{e-L}. \quad (6)$$

3. Calculation of electron Green's functions

The double time single particle electron Green's functions are calculated by equations of motion method of Zubarev [13]. The electron Green's functions are defined as

$$\begin{aligned} A(k \uparrow, \omega) &= \langle \langle c_{1k,\uparrow}^\dagger; c_{1k,\uparrow}^\dagger \rangle \rangle_\omega, \\ A(k \downarrow, \omega) &= \langle \langle c_{1k,\downarrow}^\dagger; c_{1k,\downarrow}^\dagger \rangle \rangle_\omega, \\ B(k \uparrow, \omega) &= \langle \langle c_{2k,\uparrow}^\dagger; c_{2k,\uparrow}^\dagger \rangle \rangle_\omega, \\ B(k \downarrow, \omega) &= \langle \langle c_{2k,\downarrow}^\dagger; c_{2k,\downarrow}^\dagger \rangle \rangle_\omega. \end{aligned} \quad (7)$$

The Green's functions are calculated and written as

$$\begin{aligned} A(k \uparrow, \omega) &= \frac{1}{2\pi(\omega - E_1)} \\ A(k \downarrow, \omega) &= \frac{1}{2\pi(\omega - E_2)} \\ B(k \uparrow, \omega) &= \frac{1}{2\pi(\omega - E_3)} \\ B(k \downarrow, \omega) &= \frac{1}{2\pi(\omega - E_4)} \end{aligned} \quad (8)$$

The quasiparticle energies are given by

$$\begin{aligned} E_1 &= \varepsilon_0(k) - \mu + Ge + \frac{JM}{2}, \\ E_2 &= \varepsilon_0(k) - \mu + Ge - \frac{JM}{2}, \\ E_3 &= \varepsilon_0(k) - \mu - Ge + \frac{JM}{2}, \\ E_4 &= \varepsilon_0(k) - \mu - Ge - \frac{JM}{2} \end{aligned} \quad (9)$$

4. Calculation of Magnetization (M) and Jahn-Teller distortion

The magnetization of the manganite system is defined as

$$M = N\tilde{\mu} \sum_{\alpha} (n_{\alpha\uparrow} - n_{\alpha\downarrow}), \quad (10)$$

where N , $\tilde{\mu}$ are the number of atoms per unit volume and magnetic moment of the manganese atom respectively.

The number operators $n_{1\uparrow}, n_{1\downarrow}, n_{2\uparrow}$ and $n_{2\downarrow}$ are defined by

$$\begin{aligned} n_{1\uparrow} &= \sum_k \langle c_{1k\uparrow}^\dagger c_{1k\uparrow} \rangle, \\ n_{1\downarrow} &= \sum_k \langle c_{1k\downarrow}^\dagger c_{1k\downarrow} \rangle, \\ n_{2\uparrow} &= \sum_k \langle c_{2k\uparrow}^\dagger c_{2k\uparrow} \rangle, \\ n_{2\downarrow} &= \sum_k \langle c_{2k\downarrow}^\dagger c_{2k\downarrow} \rangle. \end{aligned} \quad (11)$$

The number operators n_{is} ($i = 1, 2$) are found to be

$$\begin{aligned} n_{1\uparrow} &= \sum_k \frac{1}{\exp(E_1/k_B T) + 1}, \\ n_{1\downarrow} &= \sum_k \frac{1}{\exp(E_2/k_B T) + 1}, \\ n_{2\uparrow} &= \sum_k \frac{1}{\exp(E_3/k_B T) + 1}, \\ n_{2\downarrow} &= \sum_k \frac{1}{\exp(E_4/k_B T) + 1} \end{aligned} \quad (12)$$

The reduced magnetization $m = M/(N\tilde{\mu})$ is found to be

$$m = \sum_k [f(\beta E_1) - f(\beta E_2) + f(\beta E_3) - f(\beta E_4)], \quad (13)$$

where $b = (k_B T)^{-1}$ and the Fermi function in general is given by $f(y) = \frac{1}{e^y + 1}$. The sum over the momenta of the electrons in the conduction band is replaced by the integration over energy of the electron $\varepsilon_0(k)$ with integration limit from $-W/2$ to $+W/2$ as

$$\sum_k = \int_{-W/2}^{+W/2} N(\varepsilon_0) d\varepsilon_0.$$

Here, $N(\varepsilon_0)$ is the model density of states [14] and $2D$ is the total band width W of the conduction band. To simulate the strong energy dependence of $N(\varepsilon_0)$ around the centre of the band we take

$$N(\varepsilon_0) = N(0) \left[1 - \ln \left| \frac{D^2}{\varepsilon_0^2} \right| \right] \quad (14)$$

where $N(0)$ is unperturbed density of state of the free electron and ε_0 is the kinetic energy of the conduction band.

The chemical potential of the electron system changes with temperature (T) and the concentration of the doped impurity (x) in the system $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$. Since the impurity produces holes in the MnO_3 plane, the chemical potential (m) is determined from the average number of electrons ($n-x$) which is given by

$$(n-x) = \sum_{k,\sigma} [\langle c_{1k,\sigma}^\dagger c_{1k,\sigma} \rangle + \langle c_{2k,\sigma}^\dagger c_{2k,\sigma} \rangle]. \quad (15)$$

Using the expressions given in the eq. (12), this reduces to

$$(n-x) = \sum_k \sum_{i=1}^4 f(\beta E_i), \quad (16)$$

where the Fermi functions for different quasiparticle energies (E_i) are written earlier.

The static band Jahn-Teller distortion energy is given by

$$E_{JT} = Ge_0 \sum_{k,\sigma} [\langle c_{1k,\sigma}^\dagger c_{1k,\sigma} \rangle - \langle c_{2k,\sigma}^\dagger c_{2k,\sigma} \rangle], \quad (17)$$

where G is the strength of the electron-lattice interaction and e_0 is the static lattice distortion. The reduced lattice distortion (\tilde{e}) is given by

$$\tilde{e} = \sum_k \left| \sum_{i=1}^4 f(\beta E_i) - \sum_{j=3}^4 f(\beta E_j) \right|. \quad (18)$$

The physical quantities involved in the calculations are the band energy ($\varepsilon_0(k)$), chemical potential (m), the strength of the electron-lattice interaction (G), the lattice distortion (e), the effective interaction energy (J), the temperature (T), and the conduction band width (W). These quantities are scaled with respect to the band energy (W). The dimensionless parameters are given by

$$\begin{aligned} y &= \frac{\varepsilon_0(k)}{W}, \quad t = \frac{k_B T}{W}, \quad g = \frac{Ge_0}{W}, \quad \sim = W, \\ g_1 &= \frac{JN\tilde{\mu}}{W}, \quad \tilde{e} = \frac{e}{e_0}, \quad m = \frac{M}{N\tilde{\mu}}. \end{aligned} \quad (19)$$

5. Results and discussion

There exists ferromagnetism in manganite system $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ accompanied by Jahn-Teller lattice distortion with impurity dependant ferromagnetic Curie temperature $T_c(x) \approx 250$ K. There occurs a large drop in resistivity of the material below the Curie temperature signifying its ferromagnetic metallic character. A static Jahn-Teller distortion present in the lattice changes Curie temperature thereby changing a resistivity drop in the external doping concentration of range $0.2 \leq x \leq 0.5$. In the present work we report a simple microscopic model to study the interplay of spontaneous magnetization (M) due to ferromagnetism and lattice strain (e) due to static band Jahn-Teller distortion in the absence of external magnetic field (B). Different impurity concentrations (x) are given to the system by varying the chemical potential at various temperatures.

It is noticed that the spontaneous magnetization (M) in eq. (10) the impurity concentration (x) in eq. (15) and the lattice strain (e) in eq. (18) are coupled to each other in the form of integral equations. To study the co-existence of magnetization and lattice strain, we solve these equations numerically and self-consistently for different band fillings. The physical quantities involved in the calculations are made dimensionless with respect to the conduction band energy (W). The dimensionless parameters are magnetization (m), lattice strain (\tilde{e}), lattice coupling strength (g), magnetic coupling strength (g_1), chemical potential (z) and reduced temperature (t). It is to note that Figure 1 gives the individual plot of $m \sim t$ and $\tilde{e} \sim t$ whereas Figures 2–8 show the self-consistent plots.

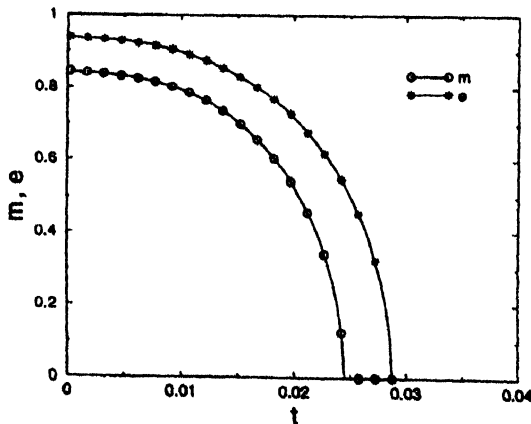


Figure 1. Individual plot of m vs t and \tilde{e} vs t for fixed values of $g = 0$, $g_1 = 0.13995$, $z = 0$ and $g = 0.074$, $g_1 = 0$, $z = 0$.

Figure 1 shows the individual plots of $m \sim t$ and $\tilde{e} \sim t$ in the absence of the magnetic field and chemical potential. Initially the magnetic coupling strength is kept constant at $g_1 = 0.13995$ in order to give a transition temperature $t_c \approx 0.024$ corresponding to Curie temperature $T_c = 240$ K.

Similarly the lattice coupling strength $g = 0.074$ gives a lattice distortion temperature $t_d \approx 0.0285$ corresponding to a distortion temperature $T_d = 285$ K. It is seen that $t_c < t_d$ initially. Then the magnetisation (m) and strain (\tilde{e}) are solved self-consistently under the half filling band situation. From Figure 2 it is observed that $t_c > t_d$ with $t_c \approx 0.029$ and $t_d \approx 0.0225$. It is also interesting to observe that the magnetization is also suppressed considerably from the initial value $m \approx 0.84$ to 0.47 due to the presence of lattice strain

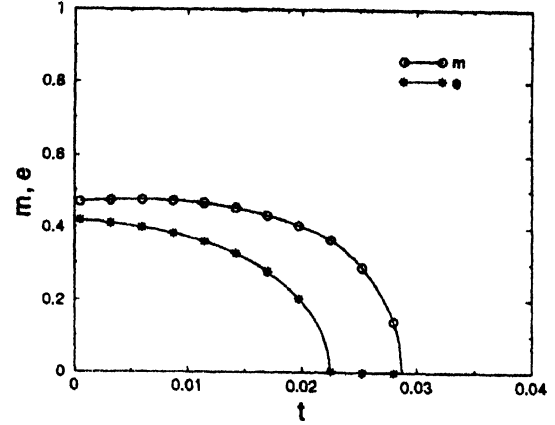


Figure 2. Self-consistent plot of m vs t and \tilde{e} vs t for fixed values of $g = 0.074$, $g_1 = 0.13995$ and $z = 0$.

Similarly the lattice strain is also suppressed considerably from its initial value $\tilde{e} \approx 0.94$ to 0.42 . In co-existence phase both the magnetization and the strain are so suppressed that the strain curve lies below the magnetization curve in contrast to its original value in individual plots. The temperature dependance of both magnetization and strain exhibit second order phase transition with respect to temperature. Experimental observations [15,16] show that the resistivity drastically reduces below Curie temperature indicating an insulating phase changing to ferromagnetic metallic state below t_c . Here in the present case the presence of Jahn-Teller distortion in the system pushes the Curie temperature to higher temperatures where the ferromagnetic metallic state is thereby extended to the higher temperatures.

Figure 3 and Figure 4 show the effect of lattice coupling strength (g) on magnetization (m) and lattice strain (\tilde{e}) respectively in their co-existence phase. As the lattice coupling increases from $g = 0.072$ to 0.075 the spontaneous magnetization (m) increases accompanied by an enhancement of the Curie temperature from 0.0265 to 0.030 as shown in the Figure 3. It signifies that the ferromagnetic metallic state is enhanced due to the presence of lattice strain. Figure 4 shows the plot of $\tilde{e} \sim t$ for different values of lattice coupling strength (g). It is observed that the lattice strain $\tilde{e}(0)$ at 0 K remains constant as the lattice coupling changes, but the lattice distortion temperature decreases with the increase of lattice coupling strength (g).

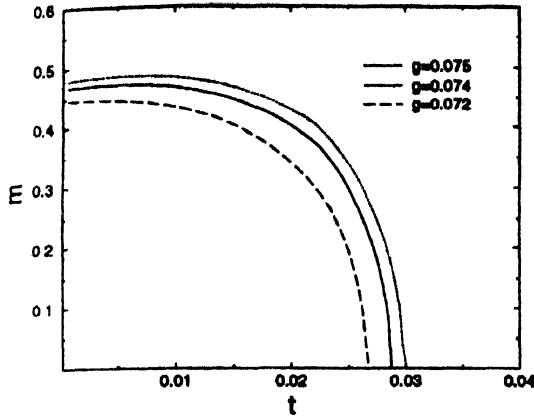


Figure 3. Plot of m vs t for different values of $g = 0.075, 0.074, 0.072$ and for fixed values of $g_1 = 0.13995, z = 0$.

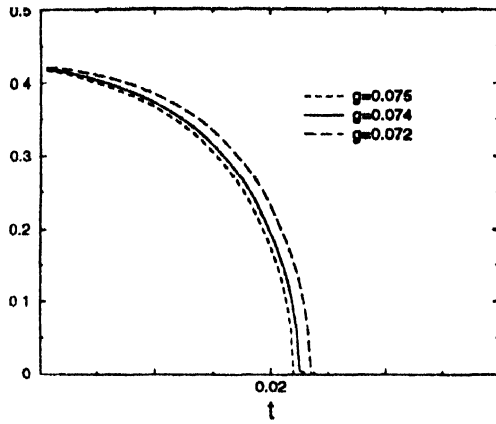


Figure 4. Plot of $\tilde{\epsilon}$ vs t for different values of $g = 0.075, 0.074, 0.072$ and fixed values of $g_1 = 0.13995, z = 0$.

Figure 5 shows the plot of $m \sim t$ for different values of ferromagnetic coupling (g_1). It is observed that the spontaneous magnetization $m(0)$ at $t = 0$ as well as the Curie temperature (t_c) remain unaffected with the change of ferromagnetic coupling (g_1).

However, the spontaneous magnetization (m) decreases with the increase of ferromagnetic coupling (g_1) in the intermediate temperature range lying between $t = 0$ and t_c . Figure 6 shows the plot of $\tilde{\epsilon} \sim t$ for different values of ferromagnetic coupling (g_1). It is seen that the lattice strain increases with the increase of ferromagnetic coupling (g_1) and the lattice distortion temperature (t_d) is enhanced as well.

Figure 7 shows the plot of $m \sim t$ and $\tilde{\epsilon} \sim t$ for various doping concentrations $x = 0.00, 0.10, 0.15$. The temperature dependence of chemical potential is such that it allows only a maximum concentration of $x = 0.15$ for stability of the model system considered here in absence of the magnetic field and the Coulomb correlation energy. It is anticipated that higher doping concentration will definitely stabilize the

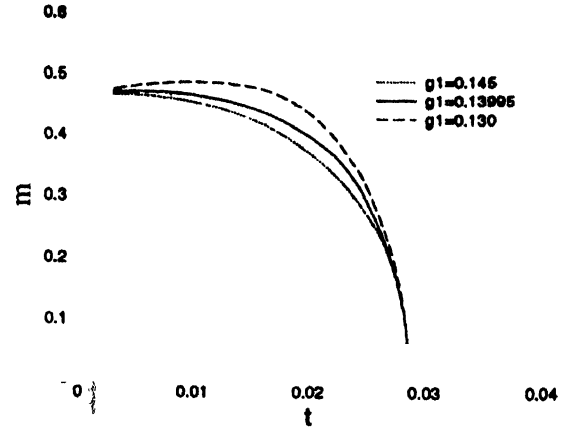


Figure 5. Plot of m vs t for different values of $g_1 = 0.145, 0.13995, 0.130$ and fixed values of $g = 0.074, z = 0$.

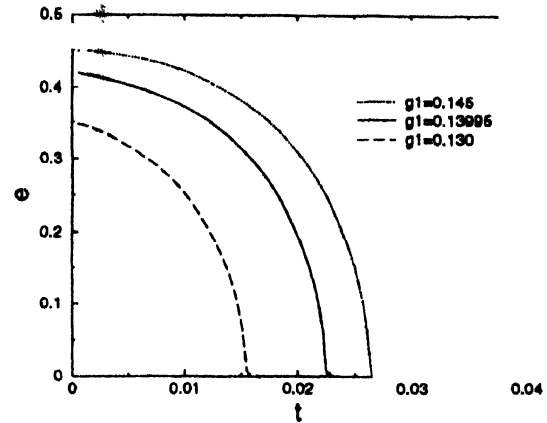


Figure 6. Plot of $\tilde{\epsilon}$ vs t for different values of $g_1 = 0.145, 0.13995, 0.130$ and fixed values of $g = 0.074, z = 0$.

system in presence of external magnetic field and the stronger Coulomb correlation. It needs further investigation.

It is seen that the lattice strain ($\tilde{\epsilon}$) and lattice distortion temperature (t_d) decrease with increase of impurity concentration. It is also seen that there is a decrease of spontaneous magnetization (m) and a reduction in Curie temperature (t_c) due to the weakening of exchange interaction (g_1) with increase of impurity concentration at higher temperatures. The higher the temperature the greater the relative effect of the exchange weakening and towards t_c doping actually causes a reduction in magnetization. However, the magnetization increases with increase of impurity concentration at lower temperatures. The opposing behaviour occurs at a critical temperature $t^* = 0.009$.

Figure 8 shows the plot of $m \sim t$ with increase of impurity concentration (x) for three different sets of parameters. The critical temperature (t^*) as discussed with respect to Figure 7 increases to a higher temperature as the elastic coupling strength (g) increases but t^* decreases when magnetic coupling strength (g_1) increases.

We conclude that the spontaneous magnetization (m) and the ferromagnetic Curie temperature (t_c) can be varied

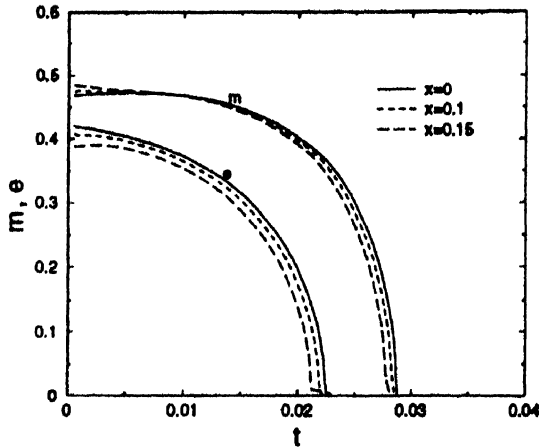


Figure 7. Plot of m vs t and \tilde{e} vs t for different values of doped concentration $x = 0, 0.1, 0.15$ and fixed values of $g = 0.074$, $g_1 = 0.13995$.

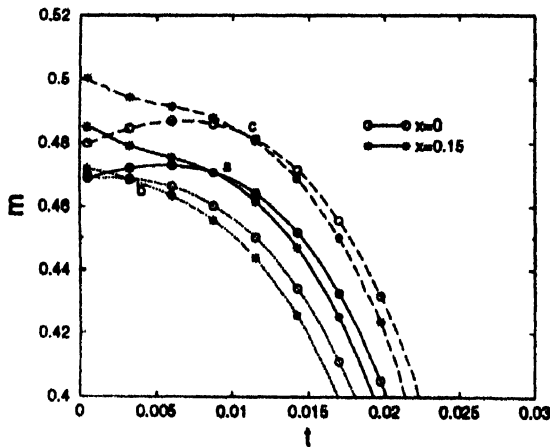


Figure 8. Plot of m vs t for different values of doped concentration $x = 0, 0.15$ and fixed values of (a) $g = 0.074$, $g_1 = 0.13995$, (b) $g = 0.074$, $g_1 = 0.145$ and (c) $g = 0.075$, $g_1 = 0.13995$.

by changing the lattice coupling, ferromagnetic coupling and impurity concentrations. The effects of the Coulomb correlation of e_g electrons and the external magnetic field are studied and are to be reported elsewhere.

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